STUDY OF DELAYED NEUTRON DECAY CURVES FROM THERMAL NEUTRON INDUCED FISSION OF ^{235}U AND ^{239}PU

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Abstract – The results of measurements of the delayed neutron decay curves obtained from the thermal-neutron induced fission of ^{235}U and ^{239}Pu are presented. The data were obtained by the periodical irradiation method using the pulsed reactor IBR-2 during the time interval 5 to 730 ms following irradiation. A comparison of these newly measured decay curves with the curves calculated using several standard delayed neutron sets was then performed. Based on these measurements, a new 7-group delayed neutron model is proposed.

INTRODUCTION

It is well known that the time dependence of the number of fission delayed neutrons, $n_d(t)$, emitted after infinite irradiation as a result of *b*-decay of various fission products, known as precursors, can be represented as a sum of exponentials:

$$n_d(t) = \sum_i a_i e^{-l_i t} \tag{1}$$

where the decay constant l_i is equal to $ln2/T_{1/2,i}$ where $T_{1/2,i}$ is the half-life of the i^{th} precursor, and the delayed neutron abundance $a_i = Y_{ci} P_n$ where Y_{ci} is the cumulative yield and P_{ni} is the probability of emission of a delayed neutron during a *b* - decay. The sum is over all delayed neutron precursors.

Because the number of delayed neutron precursors is very large (at present more than 270 are known), Eq. (1) is usually approximated by lumping precursors with similar half-lives into a smaller number of groups (i.e., a *few-group* model). The most widespread few-group model is the 6-group model first introduced by Keepin et al. (1957 and 1965). Some authors in the past have proposed a larger number of

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groups in the few-group model (e.g., Maksyutenko, 1971, and Manevich et al., 1988), however these models have never been adopted. More recently, Spriggs and others (Spriggs, 1997; Spriggs et al., 1998) proposed an 8-group model which predicts the same time-dependent behavior as predicted by a given 4-, 5-, or 6-group model. The 8-group model offers a more natural distribution of decay constants since the group half-lives correspond to the precursors with the highest yield. However, as seen in Fig. 1, it is not always clear which isotopes are dominant in a particular half-life time regime.

The most often used measurement technique employed to measure delayed neutrons requires moving a sample from the irradiation system to the detector system. This transfer usually requires a transfer time of approximately 1 second, which makes it very difficult to resolve the short-lived delayed neutrons emitted during the first second after irradiation. This initial void in the data could possibly explain the large errors and dispersion of the various results (15-35%) for the 5th and 6th groups parameters as measured by different experimenters (Tuttle, 1979; Waldo, 1981).

In this work, we discuss the periodic irradiation technique. This technique is an improvement over some of the more commonly used techniques in that it allows us to resolve the short-lived precursors during the time interval of several milliseconds to 1 second. The results of these measurements, coupled with a detailed theoretical analysis of several different fissioning systems, have allowed us to develop a new 7-group model. This model, along with the traditional 6-group model and the newly proposed 8-group model, are compared to the experimental data.

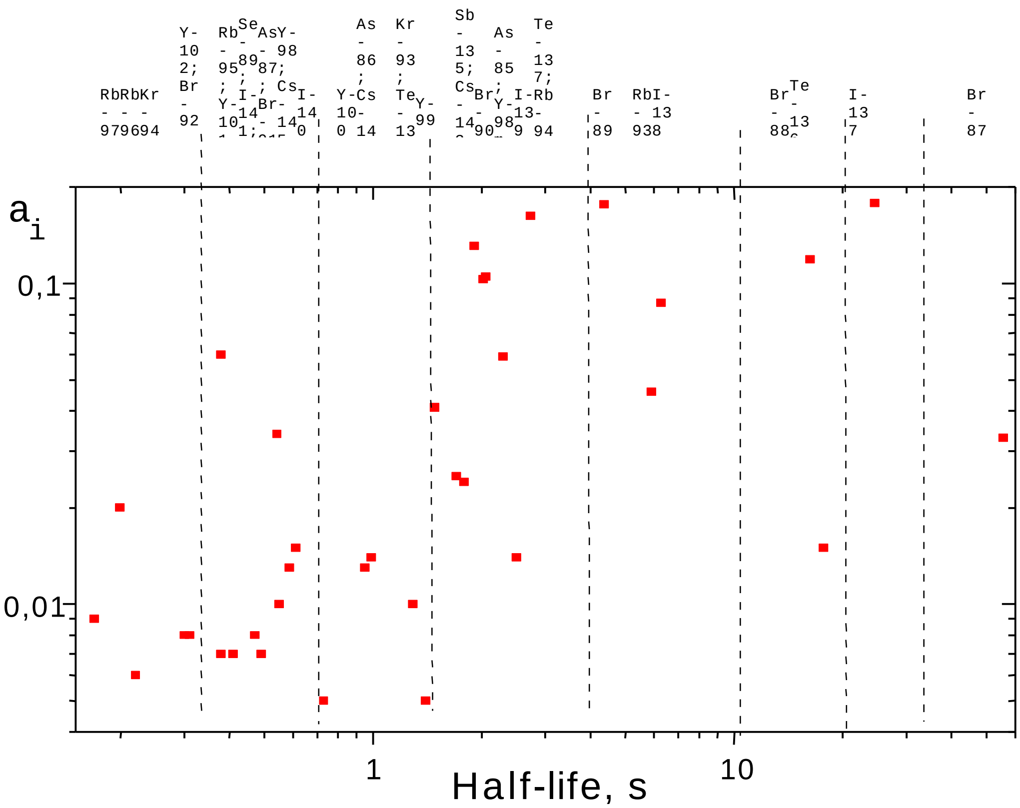

Fig. 1. Delayed neutron yield (in percent) for various precursors produced by the thermal-neutron induced fission of ^{235}U.

EXPERIMENT

The pulsed reactor IBR-2 and the *Isomer* apparatus were used to measure delayed neutron yields from thermal-neutron induced fission of ^{235}U and ^{239}Pu samples (Borzakov et al., 1994; 1995; 2000). The IBR-2 reactor (Shabalin, 1979) is a continuous-pulse reactor system operating at a pulse frequency of 5 Hz, a peak power in each pulse of 1350 MW, and an average power of 2 MW. These operating characteristics make the IBR-2 reactor a suitable neutron source for studying short-lived delayed neutrons. The main elements of the *Isomer* apparatus are (1) a chopper that produces a well-defined, thermal neutron flux at the sample, and (2) a high-efficiency detector system consisting of twelve ^{3}He counters in a moderating material. The neutron background at the *Isomer* apparatus produced by the IBR-2 reactor is minimized by physically locating the apparatus at the exit of a bent-mirror guide.

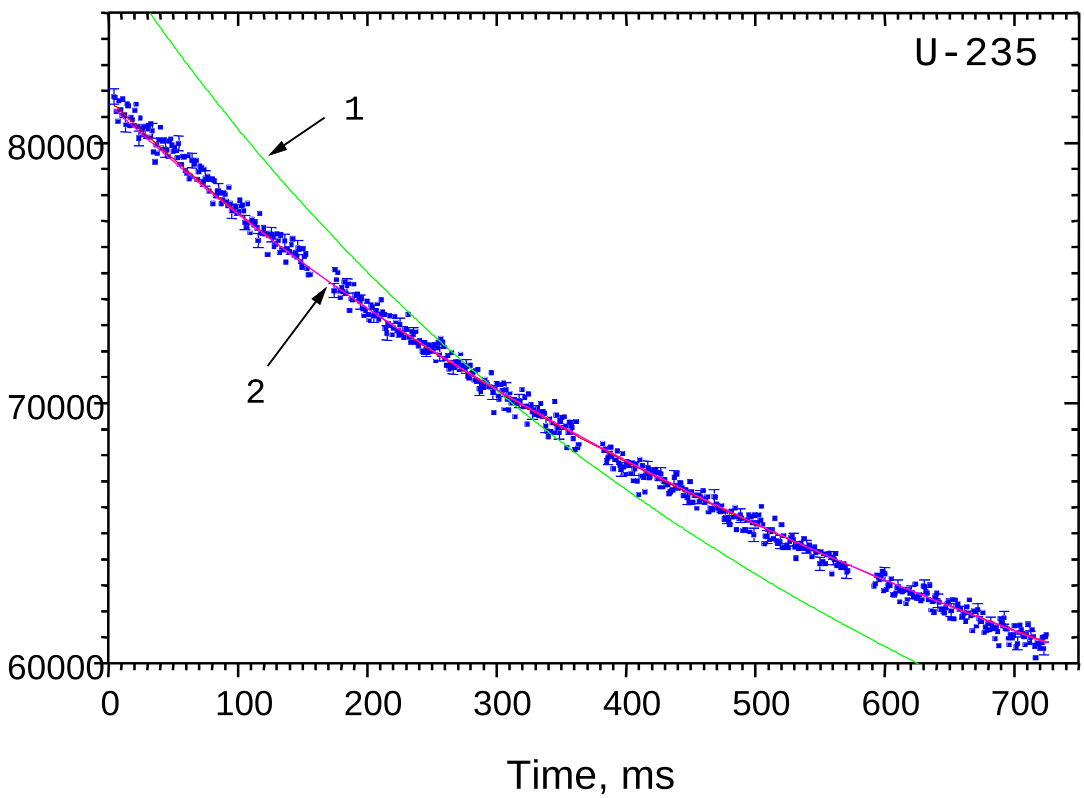

Fig. 2. The experimental data obtained for ^{235}U. The curves were calculated using the following sets of delayed neutron parameters: Curve 1 - Brady, England (1989); Curve 2 - Keepin (1957). The curves calculated using the parameters of Mills (1992), Spriggs et al. (1998), and the 7-group model presented in this work coincides with Curve 2."

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During this experiment, delayed neutron measurements for ^{235}U and ^{239}Pu samples (7 g and 10 g, respectively) were performed using the periodic irradiation technique at an irradiation frequency of 1.25 Hz. At this low frequency, the reactor undergoes three pulses during one complete revolution of the chopper. The data collected during each pulse were eliminated, causing the appearance of three gaps in our experimental data (see Fig. 2). This low chopper frequency also allowed us to measure delayed neutrons during the time interval of 5 ms to 730 ms, which was longer than previous experiments performed by us (Borzakov et al., 2000).

Since we wanted to achieve high statistical accuracy for each decay curve, the sample sizes we used were relatively large. This, however, precluded us from determining the absolute number of fissions occurring in the samples because of self-shielding effects. The data was accumulated for 40 hours for ^{235}U and 86 hours for ^{239}Pu. This produced a statistical accuracy for each count of 0.3-0.4 % for ^{235}U and ^{239}Pu in each time channel, which was set at 1.024 ms.

The neutron background during the irradiation was measured using a cadmium filter in the beam. It was found that the background did not exceed 1% of the maximum neutron count obtained during the counting interval. The decay curves obtained during these measurements, corrected for background, are shown in Fig. 2 for ^{235}U and in Fig. 3 for ^{239}Pu.

ANALYSIS AND DISCUSSION

The decay curves shown in Fig. 2 were calculated using the results of earlier published works. The delayed neutron parameters were taken from Keepin (1957), Mills et al. (1992), and Brady and England (1989) for ^{235}U, and Keepin (1957) and Brady and England (1989) for ^{239}Pu. Each curve was generated using the periodic irradiation formula,

$$\bar{n}_d(t) = N_f \cdot n_d \cdot e_d \cdot \sum_{i=1}^{m} \frac{\tilde{a}_i}{\Delta t} \cdot \frac{1-\exp(-l_i \cdot \Delta t)}{1-\exp(-l_i \cdot T)} \cdot \exp(-l_i \cdot t) \quad , \tag{2}$$

where N_f is the number of fission events, ν_d is the delayed neutron yield, T is the period of irradiation, Dt is time of irradiation within one period (Dt = 0.035 sec), e_d is the efficiency of the detector, and $\tilde{a}_i$ − is the relative weight of the i^{th} group (by definition, $\sum \tilde{a}_i = 1$).

The constant $C=N_f\, n_d\, e_d$ was determined by least-squares fit using the program MINUIT (James and Ross, 1989). As can be seen from the ^{235}U data shown in Fig. 2, the contribution of the short-lived precursors is overestimated by the delayed neutron parameters of Brady and England (1989). The delayed neutron parameters of Keepin et al. (1957) and Mills et al. (1992), however, show very good agreement with the experimental data (i.e., χ^2 = 1.3 for Keepin, and χ^2 =1.2 for Mills).

We have also compared the 8-group model of Spriggs et al. (1998) with our experimental data. In the 8-group model, the same half-lives were used for all fissionable isotopes. The expediency of this approach seems reasonable, especially for the first four groups of precursors (see Fig. 1). However, based on a calculation of the theoretical yields of the short-lived precursors using the half-lives and P_n values reported by Rudstam (1993a, 1993b), and the cumulative yields of Wahl (1988), we noted that there were very few precursors with observable relative yields greater than 0.01% in the half-life interval $T_{1/2}$ = 0.6-1.4 sec. Consequently, we decided to exclude group 8 from our delayed neutron model and simply represent the decay curve with seven groups rather than eight (see Table 1).

In the 7-group model, the average half-life for each group is defined by

$$\langle T \rangle = \frac{\ln 2}{\langle l \rangle} \quad , \tag{3}$$

$$\langle l \rangle = \sum_{i}^{j} a_i \, l_i \cdot \left(\sum_{i}^{j} a_i \right)^{-1} \quad , \tag{4}$$

where j is the number of precursors falling into a given group.

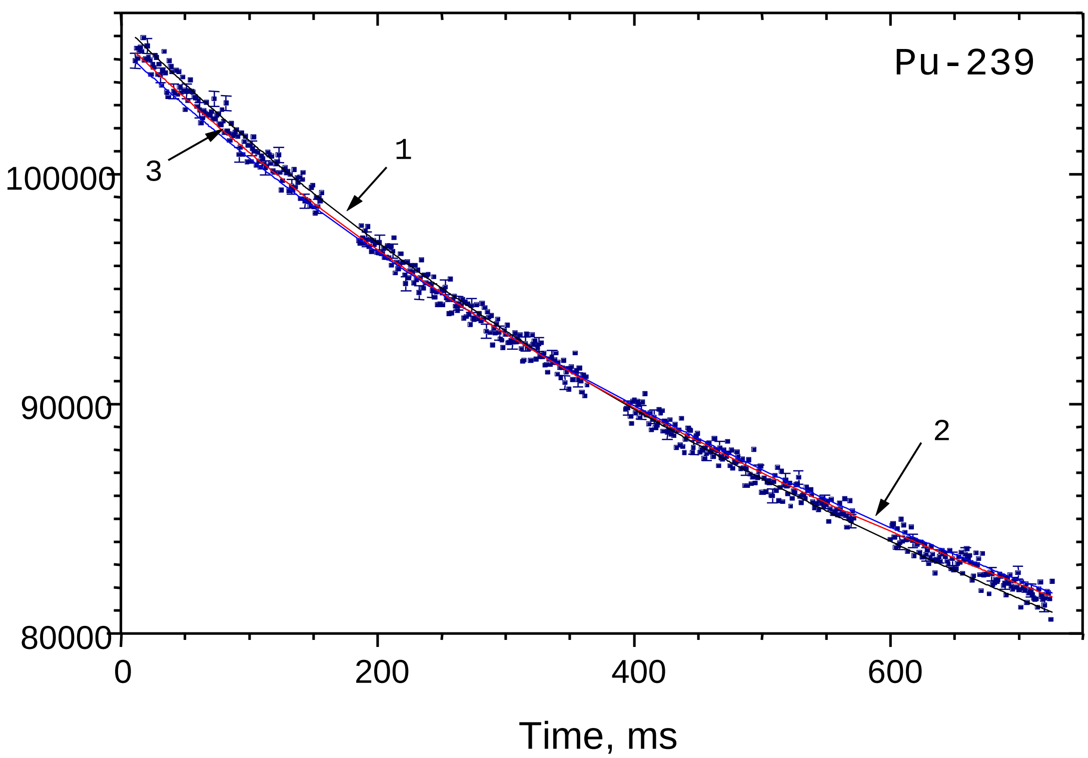

Fig. 3.   The experimental data obtained with the sample ^{239}Pu. The curves were calculated using the following sets of parameters: Curve 1- Keepin (1957); Curve 2 - Brady-England (1989); Curve 3 – seven-group model presented in this work.

As can been seen from Tables 1 and 2, the parameters of the first 4 groups don't change considerably from the half-lives chosen in the 8-group model of Spriggs et al. (1998).

Table 1. Seven-Group Parameters for Thermal Fission of ^{235}U.

Group number	Precursor	$T_{1/2,i}$ sec	a_i (per 100 fissions)	Average $T_{1/2,j}$, sec	Group yield (per 100 fissions)
1	Br-87	55.6	0.054	55.6	0.054
2	I-137	24.5	0.233	24.5	0.233
3	Te-136	17.7	0.015	16.4	0.134
	Br-88	16.23	0.119		
4	I-138	6.27	0.087	4.99	0.309
	Rb-93	5.91	0.046		
	Br-89	4.37	0.176		
5	Rb-94	2.73	0.162	2.07	0.664
	Te-137	2.50	0.014		
	I-139	2.29	0.059		
	Y-98m	2.05	0.105		
	As-85	2.02	0.103		
	Br-90	1.91	0.131		
	Cs-143	1.79	0.024		
	Sb-135	1.70	0.025		
	Y- 99	1.48	0.041		
	Kr-93	1.29	0.010		
6	Cs-144	0.99	0.014	0.494	0.159
	As-86	0.95	0.013		
	Y-100	0.73	0.005		
	I-140	0.61	0.015		
	Cs-145	0.586	0.013		
	Y-98	0.549	0.010		
	Br-91	0.542	0.034		
	As-87	0.490	0.007		
	I-141	0.470	0.008		
	Se-89	0.410	0.007		
	Y-101	0.38	0.007		
	Rb -95	0.379	0.060		
7	Br-92	0.31	0.008	0.218	0.045
	Y-102	0.30	0.008		
	Kr-94	0.22	0.006		
	Rb-96	0.199	0.020		
	Rb-97	0.169	0.009		

Table 2. Seven-Group Parameters for Thermal Fission of ^{239}Pu

Group number	Precursor	$T_{1/2,i}$ (sec)	a_i (per 100 fissions)	Average value $T_{1/2,j}$ (sec)	Group yield (per 100 fissions)
1	Br-87	55.6	0.017	55.6	0.017
2	I-137	24.5	0.157	24.5	0.157
3	Te-136	17.7	0.009	16.5	0.043
	Br-88	16.23	0.034		
4	I-138	6.27	0.061	5.40	0.123
	Rb-93	5.91	0.019		
	Br-82	4.37	0.043		
5	Nb-105	2.95	0.0117	2.097	0.248
	Rb-94	2.73	0.0635		
	I-139	2.29	0.0297		
	Y-98m	2.05	0.0440		
	As-85	2.02	0.0165		
	Br-90	1.91	0.0333		
	Cs-143	1.79	0.0103		
	Sb-135	1.70	0.0117		
	Y-99	1.48	0.0270		
6	Cs-144	0.99	0.0044	0.460	0.061
	Y-100	0.73	0.0034		
	I-140	0.61	0.0031		
	Cs-145	0.586	0.0032		
	Y-98	0.549	0.0085		
	Br-91	0.542	0.0046		
	Y-101	0.380	0.0027		
	Rb-95	0.379	0.0310		
7	Rb-96	0.199	0.0070	0.191	0.00915
	Rb-97	0.169	0.00215		

When compared to the experimental data, we note that the 7-group model data for ^{235}U shows good agreement (i.e., $\chi^2 = 1.4$). The 7-group decay curve practically coincides with the curve 2 on the Fig. 2.

The experimental data obtained for ^{239}Pu were analyzed using the previously discussed 6-group models and the proposed 7-group shown in the Table 2. The calculated curves are compared with experimental data in Fig. 3. The agreement of our 7-group model with the experimental data for ^{239}Pu is not quite as good as for ^{235}U, but, nevertheless, still shows good agreement (i.e., $\chi^2 = 2.6$). In comparison, the decay curves calculated using the Brady-England parameters and the Keepin parameters yield $\chi^2 = 2.3$ and $\chi^2 = 1.8$, respectively.

CONCLUSIONS

The measured delayed neutron decay curves for the thermal-induced fission of ^{235}U and ^{239}Pu during the time interval 5 to 730 ms after irradiation are presented in this work. These measurements were performed using the periodic irradiation technique, which allowed us to obtain high statistical accuracy (i.e., 0.3-0.4 % per 1.024 ms channel width). A comparison of the experimental data with decay curves calculated using various 6-, 7-, and 8-groups models was also performed. It was determined that the decay curve for both ^{235}U and ^{239}Pu during this time interval was adequately represented using the 7-group model proposed in this work.

ACKNOWLEDGEMENT

The authors wish to thank Dr. Yu.V. Grigoriev for providing the samples used in this experiment, and Dr. W.I. Furman for his steadfast support. We are also grateful to Dr. G. D. Spriggs and Dr. V. M. Piksaikin for their useful discussion of experimental data and its interpretation. This work was supported, in part, by ISTC project #471 and grant RFFR 95-02-03740.

REFERENCES

Borzakov S. B., Dermendjiev E., Zamyatnin Yu. S., Nazarov V.M., Pavlov S. S., Rogov A. D., Ruskov I. (1994), preprint JINR P3-94-447, Dubna.

Borzakov S. B., Dermendjiev E., Zamyatnin Yu. S., Nazarov V. M., Pavlov S. S., Rogov A. D., Ruskov I. (1995), *Atomic Energy*, (in Russian) **79**, 231.

Borzakov S. B., Andreev A. N., Dermendjiev E., Filip A., Furman W. I., Panteleev Ts., Ruskov I., Zamyatnin Yu. S., Zeinalov Sh. (2000), *Physics of Atomic Nuclei*, **63**, 530.

Brady M. C., England T. R. (1989), *Nucl. Sci. Eng.*, **103**, 129.

James F., Ross M. (1989), CERN Program Library, D506.

Keepin G. R., T. F. Wimett, and R. K. Zeigler (1957), *Phys. Rev.*, **107**, No. 4, 1044.

Keepin G. R. (1965), Physics of Nuclear Kinetics, Addision Wesley, Reading. MA.

Maksyutenko B. V. (1971), *Sov. Nucl. Phys.*, **13**, No. 2.

Manevich L. G., Nemirovskii P. E., Yudkevich M. S. (1988), *Voprosy Atomnoi Nauki i Tekchniki*, Yadernye Constanty, **2**, 3.

Mills R. W., James M. F., and Weaver D. R. (1992), Nuclear Data for Science and Technology, Proceedings, FRG, Juelich, p. 86, Springler-Verlag, Berlin.

Rudstam G. (1993a), IAEA Report INDC (SWD) 24/L+P, Vienna.

Rudstam G. (1993b), Atomic Data and Nuclear Data Tables, **53**, 1.

Shabalin E. P. (1979), Fast Pulsed and Burst Reactors, Pergamon Press.

Spriggs G. D., Campbell J. M., Piksaikin V. M. (1998), LA-UR-98-1619, Los-Alamos.

Spriggs G. D. (1997), oral presentation at Colloquy on Delayed Neutron Data, Obninsk, Russia.

Tuttle R. J. (1975), *Nucl. Sci. Eng.*, **56**, 37.

Wahl A. C. (1988), Atomic Data and Nuclear Data Tables, **39**, 1.

Waldo R. W. et al. (1981), *Phys. Rev*. **C23**, 1113.